

## Stability of Uranium incorporated into Fe(hydr)oxide structures under fluctuating redox conditions

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Owing to ecosystem and human health consequences, understanding uranium's potential for mobility in environmental settings is important. In anaerobic soils and sediments, the oxidized form of uranium (UO<sub>2</sub><sup>2+</sup>) may be reduced through biological or chemical pathways to U(IV), forming sparingly soluble solids. However, formation of ternary calcium-carbonate complexes of uranium may limit reduction. Additionally, authogenic UO<sub>2</sub> is potentially susceptible to reoxidation in the presence of commonly occurring oxidants including molecular oxygen. Therefore, determining reaction pathways of uranium that lead to solids stable (i.e., having limited solubility) under both aerobic and anaerobic conditions is critical for limiting dissolved concentrations and migration of uranium. We examined the fate of uranium incorporated into transforming Fe(hydr)oxides under cyclic oxidizing and reducing conditions. Uranium in treatment systems containing ferrihydrite, 4 mM Ca, 3.8 mM HCO<sub>3</sub><sup>-</sup> and either 3 or 10 mM Fe(II), that were maintained under reducing conditions for 15 d followed by 5 d of oxidation (1 cycle) for 3 cycles exhibited widely diverging fates. Uranium EXAFS and XANES analysis reveal oxidized uranium both adsorbed on and incorporated in the structure of goethite and magnetite. High levels of Fe(II) and long incubation times result in conversion of incorporated U(VI) into discrete UO<sub>2</sub> phases associated, dominantly, with magnetite. Our results reveal that active transformation of Fe(III) phases can result in structural incorporation of U(VI) that is stable toward fluctuating redox conditions.

## Field evidence of relationships between plankton and <sup>210</sup>Po, <sup>210</sup>Pb

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### Unique Radionuclide Pair

The final alpha-emitting product of the radioactive decay of <sup>238</sup>U, <sup>210</sup>Po (*t*<sub>1/2</sub> 138 d), is produced by the beta decay of <sup>210</sup>Pb (*t*<sub>1/2</sub> 22 yr) via the short-lived intermediate <sup>210</sup>Bi (*t*<sub>1/2</sub> 5 d). <sup>210</sup>Po has been used to trace particle transfer, ocean circulation, and vertical flux of particulate matter in the ocean. Like uranium and thorium, radioactive lead and polonium should come to secular equilibrium in the surface ocean, but the two are often in disequilibrium [1]. Laboratory and field evidence suggests that <sup>210</sup>Po has a higher affinity for organic matter than <sup>210</sup>Pb.

### Interactions with Phytoplankton and Zooplankton

<sup>210</sup>Po and <sup>210</sup>Pb were measured on phytoplankton cells rinsed with an oxalate solution to remove surface-bound metals [2]. Previous laboratory studies suggest that approximately 50% of the <sup>210</sup>Po associated with phytoplankton cells is found inside the cytoplasm [3]. Our field experiments indicated that this result may hold true in wild populations as approximately 60% of the <sup>210</sup>Po associated with phytoplankton was not removed by the rinse. In addition, vertical profiles of zooplankton were collected during daytime and at night to investigate the vertical transport of <sup>210</sup>Po and <sup>210</sup>Pb caused by the activity of daily migrating zooplankton. A significant fraction of the total water column <sup>210</sup>Po and <sup>210</sup>Pb was found associated with the migrating zooplankton caught in the plankton recorder, and the distribution of the radionuclides was significantly different between day (high at depth) and night (high at surface) tows. Both of these experiments provide further insight into the behaviour of this unique radioisotope pair, and can be incorporated into current applications of <sup>210</sup>Po as an organic carbon tracer in the surface ocean.

[1] Cochran *et al.* (1983) *EPSL* **65**, 433–452. [2] Tovar-Sanchez *et al.* (2003) *Marine Chem.* **82**, 91–99. [3] Stewart & Fisher (2003) *L & O* **48**, 1193–1201.